Neodymium isotopic composition of the oceans: A compilation of seawater data

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ABSTRACT

A global compilation of the neodymium isotopic composition of seawater is presented. With 880 data points, it confirms the gradual ε_{Nd} increase for intermediate/deep water masses from the northwest North Atlantic, via the Austral and Indian oceans, to the Pacific. This confirms the usefulness of this tracer for studying large scale oceanic circulation. The compilation stresses the need for documenting the oceans south of 30°S, from which less than 4% of the data are derived. The associated neodymium concentrations display heterogeneous vertical gradients amongst major oceanic basins. In addition to particle remineralization along the global thermohaline circulation, the database suggests that basin size differences also contribute to that heterogeneity.

INTRODUCTION

The neodymium isotopic composition, hereafter abbreviated Nd IC, is expressed as $\varepsilon_{Nd} = \left(\frac{\left(\frac{^{143}Nd}{^{144}Nd} \right)_{sample}}{\left(\frac{^{143}Nd}{^{144}Nd} \right)_{CHUR}} - 1 \right) \times 10^4$, where CHUR stand for CHondritic Uniform Reservoir and represents a present day average Earth value $(^{143}Nd/^{144}Nd)_{CHUR}$=0.512638 [*Jacobsen and Wasserburg*, 1980]. The Nd IC is heterogeneous on the continents, ranging from ε_{Nd}=−56 in old granitic cratons to +12 in recent mid-oceanic ridge basalts (GEOROC database: http://georoc.mph-mainz.gwdg.de/georoc/), reflecting both the heterogeneous ages and Sm/Nd ratios of the geological formations. It is therefore used as a tracer of the origin of the terrigeneous matter, such as aerosols [*Grousset et al.*, 1992] and river particles [*Mearns*, 1988]. In the ocean, the Nd is of lithogenic origin. Its concentration is of the order of 10^{-12} g/g (10 pmol/kg). It is predominantly found in the dissolved form (90 to 95 %, [*Sholkovitz et al.*, 1994; *Jeandel et al.*, 1995; *Alibo and Nozaki*, 1999]). The oceanic Nd IC distribution is heterogeneous, controlled both by Nd sources and sinks, and by the oceanic circulation. Besides river and aeolian inputs, Boundary Exchange – dissolution of Nd from margin sediments associated to removal of dissolved Nd from seawater – seems to dominate Nd sources and sinks [*Tachikawa et al.*, 2003; *Lacan and Jeandel*, 2005b; *Arsouze et al.*, 2009]. The major process involved in Nd removal is considered to be adsorption onto organic and Fe-Mn oxide coatings of sedimenting particles [*Sholkovitz et al.*, 1994; *Haley et al.*, 2004]. The oceanic residence time of Nd is of the order of 300 to 1000 years [*Tachikawa et al.*, 2003; *Arsouze et al.*, 2009], which is shorter than the mean oceanic mixing time. These features have led to use the Nd IC as a tracer of oceanic circulation for the modern and past ocean [e.g. *Piepgras and Wasserburg*, 1987; *Rutberg et al.*, 2000; *Piotrowski et al.*, 2008] and dissolved/particles interactions [e.g. *Tachikawa et al.*, 1997]. Although the usefulness of Nd IC in oceanography has been recognized since the early 80s, global modeling of the Nd IC of the oceans has became possible only for a few years because of the improvement of our

understanding of the Nd oceanic cycle and of an increase of the global oceanic database [e.g. *Arsouze et al.*, 2007]. Modeling is a powerful approach to examine the sensitivity of Nd IC to the various processes that are still difficult to quantify. Model-data comparison is one of the most efficient approaches that allow better constraining Nd behavior in the marine environment. Besides, Nd IC application to paleoceanographic studies necessitates the validation that marine archives faithfully record seawater isotopic signatures.

In such a context, databases of the Nd IC of i) the lithogenic inputs to the ocean and ii) the seawater itself appeared useful. A database of the lithogenic inputs to the ocean was published a few years ago [*Jeandel et al.*, 2007]. We have maintained an up-to-date and freely available database of seawater Nd IC measurements on the LEGOS marine geochemistry group website (http://www.legos.obs-mip.fr/recherches/equipes/geomar/Nd_DataBase). In the present paper, we publish this database as of September 1^{rst} 2011, we briefly discuss the methodology and intercalibration issues and finally we detail the most significant features of this global data compilation. We believe that this compilation could be useful, notably it could be used as a reference for modeling and paleoceanographic studies.

Nd IC has been identified as a "key parameter" of the GEOTRACES program (Scientific Committee on Oceanographic Research, [*GEOTRACES Planning Group*, 2006]). The number of seawater Nd IC data should therefore significantly increase in the coming years. These new and forthcoming data, acquired in the framework of GEOTRACES labeled cruises, will be archived in the GEOTRACES International Data Assembly Centre (http://www.bodc.ac.uk/geotraces/).

METHODOLOGY

All seawater Nd IC data published up to September 2011 to our knowledge are reported here [*Piepgras et al.*, 1979; *Piepgras and Wasserburg*, 1980, 1982, 1983; *Stordal and Wasserburg*, 1986; *Piepgras and Wasserburg*, 1987; *Piepgras and Jacobsen*, 1988; *Spivack and Wasserburg*, 1988; *Bertram and Elderfield*, 1993; *Jeandel*, 1993; *Henry et al.*, 1994; *Shimizu et al.*, 1994; *Jeandel et al.*, 1998; *Tachikawa et al.*, 1999; *Amakawa et al.*, 2000; *Lacan and Jeandel*, 2001; *Lacan*, 2002; *Amakawa et al.*, 2004a, 2004b; *Lacan and Jeandel*, 2004a, 2004b, 2004c; *Tachikawa et al.*, 2004; *Vance et al.*, 2004; *Dahlqvist et al.*, 2005; *Lacan and Jeandel*, 2005a, 2005b; *Tazoe et al.*, 2007a, 2007b; *Andersson et al.*, 2008; *Amakawa et al.*, 2009; *Godfrey et al.*, 2009; *Porcelli et al.*, 2009; *Rickli et al.*, 2009; *Zimmermann et al.*, 2009a, 2009b; *Rickli et al.*, 2010; *Copard et al.*, 2011; *Tazoe et al.*, 2011]. When Nd concentration is also available, it is also reported. In contrast, studies reporting seawater Nd concentration only (without Nd IC) are not considered in this study. Seawater is defined here as waters with salinities >25. Both unfiltered and filtered sampled are included (authors usually assume that unfiltered samples well represent dissolved Nd, as particulate Nd usually accounts for only ~5% of the total; cf. introduction).

Since the early work of G.J. Wasserburg and his collaborators, various protocols have been used to measure seawater Nd IC. They are briefly summarized. They include four main steps: the sampling (and sometimes filtration), the preconcentration, the purification and the analysis of Nd isotopic ratios. Sampling has been performed using standard Niskin bottles, trace metal clean Go-Flo bottles, or various surface water sampling devices, such as inlets from the ship hulls, towed fishes or snorkels. The preconcentration step has been performed by Fe or Mn REE co-precipitation, REE complexation by a HDEHP-H_2MEHP mixture adsorbed on a C_{18} cartridge, or REE adsorption on MnO_2 fibers. One study used Nd diffusion through a layer of poly acrylamide hydrogel (DGT), in situ, allowing sampling and preconcentration to be performed simultaneously. MnO_2 fibers were sometimes also deployed in situ, also allowing combining these two steps. The purification has been performed by various ion exchange chromatographies. Finally Nd isotope ratios have been determined by multi-collector thermal ionization and inductively coupled plasma mass spectrometry.

Thanks to the recent intercalibration conducted on a world scale in the framework of the GEOTRACES program, most of these analytical procedures have been validated and a detailed review of those currently in use is given in [*van de Flierdt et al.*, submitted].

The intercalibration exercise revealed that the inter-laboratory reproducibility for dissolved ε_{Nd} measurement is 0.5 to 0.6 ε_{Nd} units (2 standard deviation)[*van de Flierdt et al.*, submitted], that is well below the natural variation range in the ocean (cf. below). Most of the laboratories having reported seawater Nd IC data took part to the Geotraces intercalibration exercise. Nevertheless, the pioneer group of G.J Wasserburg and his co-workers could not take part to this recent exercise. Therefore, we compared our own data with those published by this pioneer group. Because surface waters are more susceptible of temporal variations than intermediate and deep waters, the following comparisons are restricted to the latters. The signature of the core of the North Atlantic Deep Water first characterized by ε_{Nd}=-12.6 by Piepgras and Wasserburg [1987] (TTO/TAS St. 63, 8°N, 2910m), was then characterized by ε_{Nd}=-12.3 by Jeandel [1993] (SAVE St. 302, 33°S, 2763m). This consistency was later underlined on the basin scale [*von Blanckenburg*, 1999] and further confirmed by the GEOTRACES intercalibration data, ε_{Nd}=-12.14 [*van de Flierdt et al.*, submitted] (BATS, 32°N, 2000m). The data published by Spivack and Wasserburg [1988] in the North-eastern Atlantic for the North Atlantic Central Water and in the Gibraltar Strait for the Mediterranean Water compare also well with the values published by Tachikawa et al. [1999, 2004] (cf. Table 1 and Fig. 1). Finally, an excellent agreement was also found in the Labrador Sea Water, the Iceland Scotland Overflow Water, the Norwegian Sea Deep Water and the Greenland Sea Deep Water, when comparing Piepgras and Wasserburg data [1987] with Lacan and Jeandel data [2004a, 2004b, 2004c, 2005a], despite the fact the latter samples were taken 16 to 18 years later than the former (cf. Table 1 and Fig. 1). The good consistency between all these data made us confident to include them in the present database.

Table 1. Comparison of data from G.J Wasserburg and his co-workers with data produced at LEGOS (Toulouse, France)

Cruise, station	Depth. (m)	Salinity	ε_{Nd}	2σ	Conc. (pmol/kg)	Water mass	Ref.
Hudson 83-036 St. 11; 52.08°N -47.02°E	1500	34.872	-14.4	0.4	18.1	LSW	a
Signature/GINS St. 6; 50.20°N, -45.68°E	1650	34.86	-13.9	0.4	17.4		b
Hudson 83-036 St. 11; 52.08°N -47.02°E	2500	34.932	-13.8	0.5	16.7	NEADW	a
	3000	34.933	-13.4	0.4	17.3		a
Signature/GINS St. 6; 50.20°N, -45.68°E	2499	34.90	-13.2	0.4	18.3		b
TTO/NAS Station 142; 61.35°N -8.02°E	750	34.905	-7.7	0.6	21.4	ISOW	a
Signature/GINS St. 23; 60.50°N, -5.00°E	988	34.90	-7.3	0.4	24.6		c
TTO/NAS Station 144; 67.67°N -3.28°E	3750	34.909	-9.5	0.5	16.3	NSDW	a
Signature/GINS St. 26; 69.03°N, 7.95°E	2972	34.90	-9.8	0.4	27.3		c
TTO/NAS Station 149; 76.88°N 1.03°E	2800	34.901	-10.7	0.4	16.8	GSDW	a
Signature/GINS St. 30; 76.74°N, -2.33°E	2512 (unfiltered)	34.90	-10.8	0.4	19.6		d
TTO-TAS station 80; 27.83°N, -30°53°E	389	35.942	-11.0	0.8	13.9	NACW	e
EUMELI Station E3O; 21°N, -31°E	250	36.47	-10.5	0.2	18.6		f
	500	35.63	-10.5	0.2	18.2		f
Station MED-15; 36.07°N, -6°E	250	-	-9.9	0.5	28.0	MW	e
	400	38.517	-10.1	0.7	32.4		e
Station 5, 35.9°N, -5.65°E	350	38.48	-9.4	0.2	-		g

Cf. Fig 1 for station locations.

References: a [*Piepgras and Wasserburg*, 1987]; b [*Lacan and Jeandel*, 2005a]; c [*Lacan and Jeandel*, 2004b]; d [*Lacan and Jeandel*, 2004a]; e [*Spivack and Wasserburg*, 1988]; f [*Tachikawa et al.*, 1999]; g [*Tachikawa et al.*, 2004].

LSW: Labrador Sea Water, NEADW; North East Atlantic Deep Water; ISOW: Iceland Scotland Overflow Water; NSDW: Norwegian Sea Deep Water; GSDW: Greenland Sea Deep Water; NACW: North Atlantic Central Water; MW: Mediterranean Water.

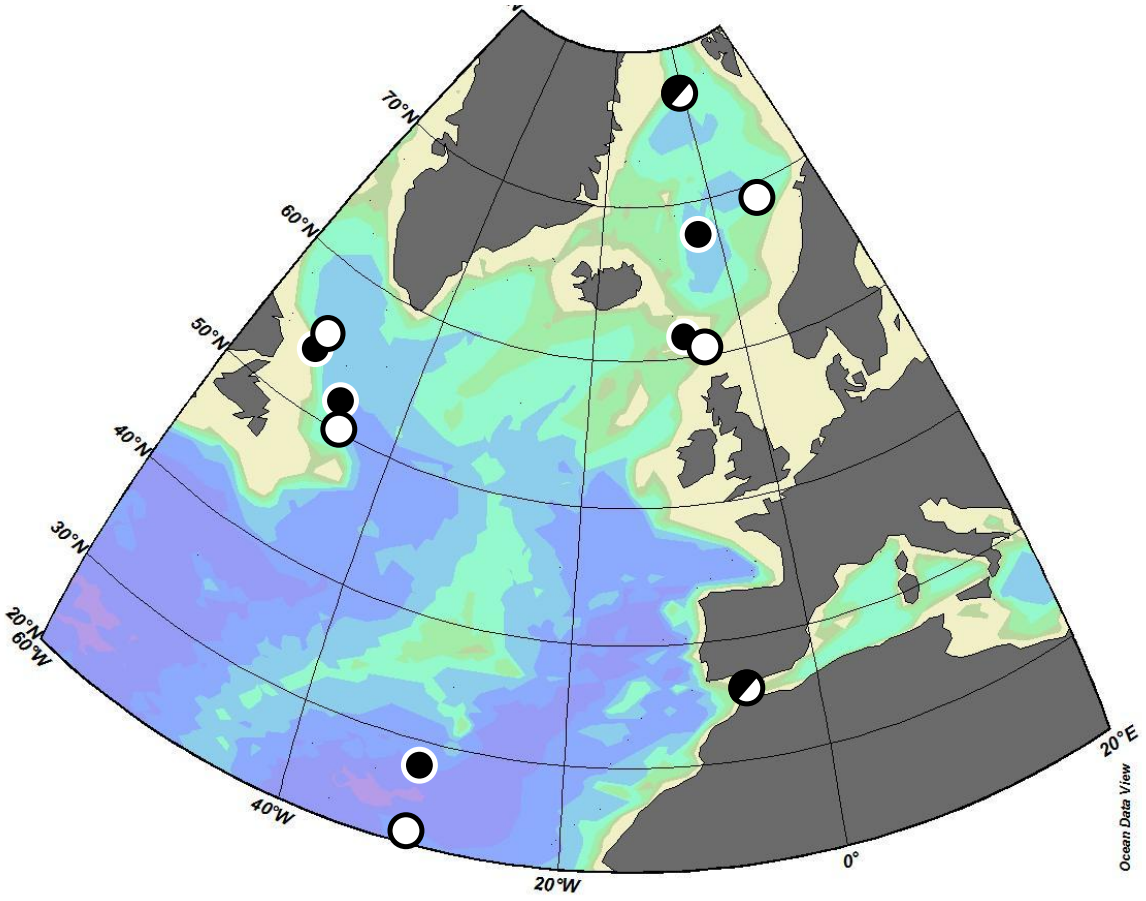
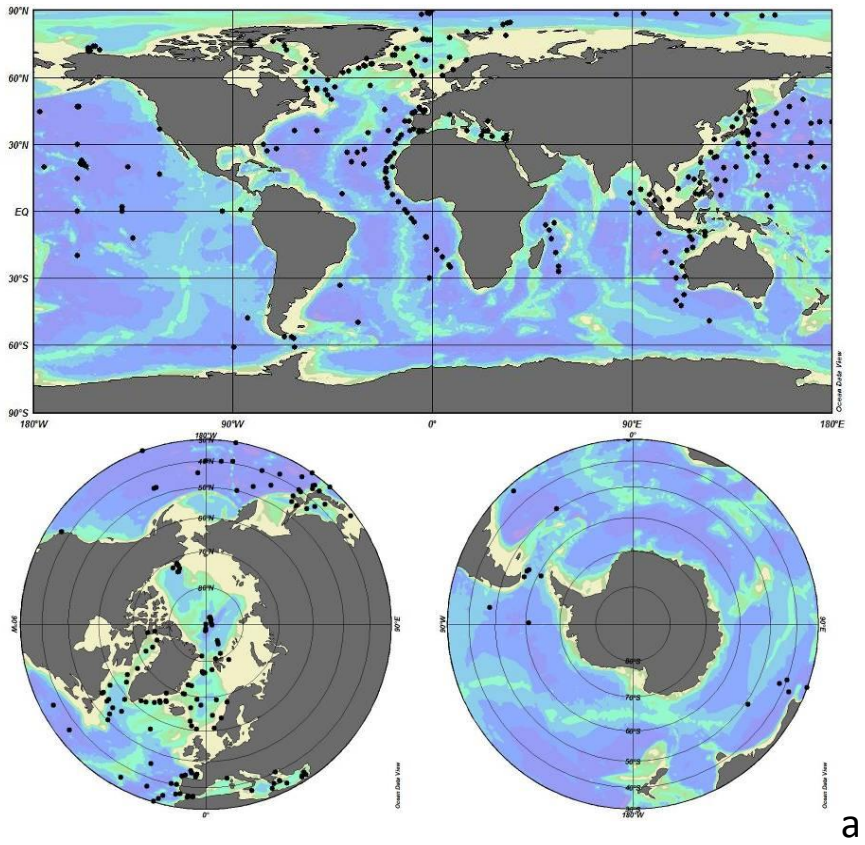
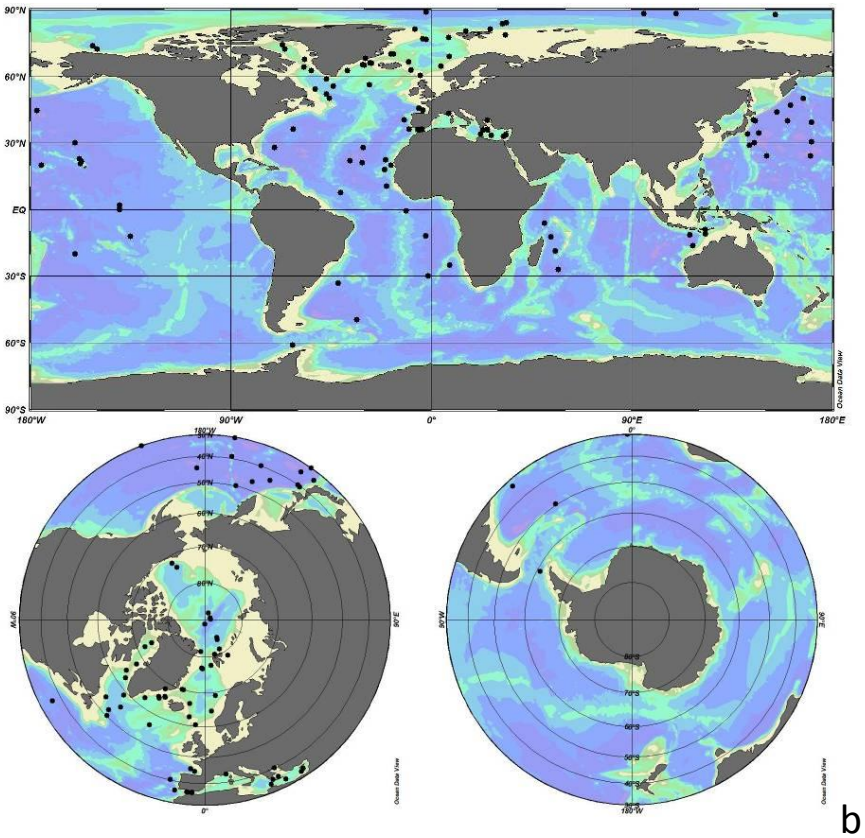


Fig. 1:
Locations of the stations used for comparing the data published by Wasserburg's group (black circles), with those published by the authors of the present work (white circles). Where 2 stations are too close to each other to be represented by 2 distinct symbols, a black and white symbol is used. Cf. Tab. 1 for ε_{Nd} data. Figure made with Ocean Data View [*Schlitzer*, 2009].



a



b

Fig 2.
a: Location of all stations with ε_{Nd} data. b: Location of stations with at least 3 ε_{Nd} data per station. Figure made with Ocean Data View [*Schlitzer*, 2009].

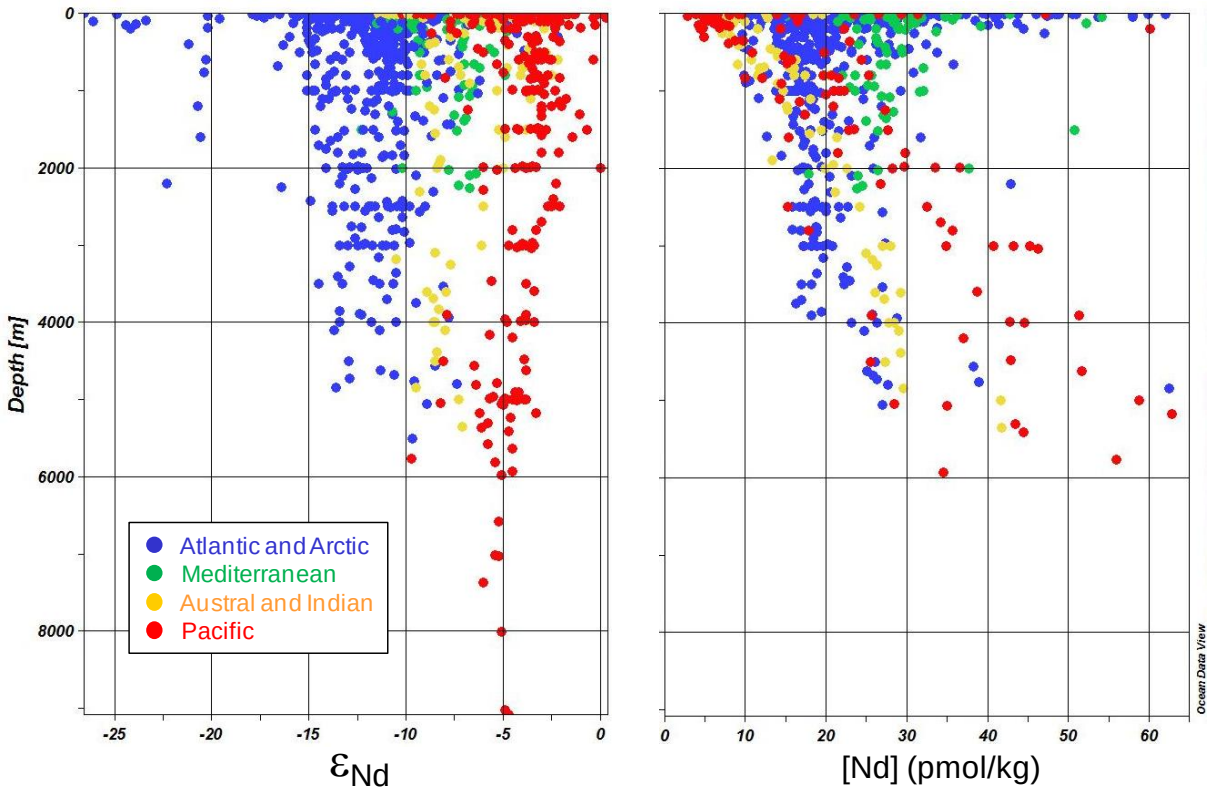


Fig. 3.
All ε_{Nd} data as a function of depth (left), and corresponding Nd concentration when documented (right). Colors indicate locations, Blue: Atlantic and Arctic; Green: Mediterranean; Yellow: Austral and Indian; Red: Pacific. The Nd concentration scale is restricted to values lower than 65 pmol/kg for clarity, excluding 13 shallow near shore data (< 2% of the total) from the North Atlantic. Figure made with Ocean Data View [*Schlitzer*, 2009].

RESULT AND COMMENTS

The database is presented in Table 2 (supplementary material). Figure 2 shows the geographical distribution of the data. The database includes a total of 880 ε_{Nd} data points, from 280 different stations, and 686 Nd concentrations. Out of these 280 stations, only 110 (i.e. 39%) include more than 3 ε_{Nd} data points (Fig. 2b). About two thirds (66%) of the ε_{Nd} data are located in the top 1000 m of the water column (Fig. 3). The global mean Nd IC is ε_{Nd}=−8.8, with values ranging from -26.6 to +0.4. The latitudinal distribution is very uneven, with 84% of the ε_{Nd} data in the Northern hemisphere, and very few data south of 30°S (less than 4%). Most data are found in the North Atlantic and North Pacific oceans, whereas the South Atlantic, the Indian, the Austral and the South Pacific oceans are very scarcely documented (see also Supplementary Fig. 1).

Figure 4 displays the Nd IC values averaged between the surface and 400m depth and between 800m depth and the bottom. Although slightly different, the two distributions display the same trends on the first order. Values are minimal in the northwest Atlantic, especially in the Baffin Bay, intermediate in the Austral and Indian oceans, and maximal in the Pacific. This is also visible in Fig. 3 and Supplementary Fig. 1. This global pattern is in good agreement with the Nd isotopic signature of the continents close to the coasts, non-radiogenic (i.e. relatively depleted in Sm borne ^{143}Nd, therefore with relatively low ε_{Nd} values) around the Atlantic Ocean and radiogenic (i.e. relatively enriched in Sm borne ^{143}Nd, therefore with relatively high ε_{Nd} values) around the Pacific [*Jeandel et*

al., 2007]. It confirms what was previously known about the global Nd isotope cycle [e.g. *Piepgras et al.*, 1979; *Goldstein and Hemming*, 2003].

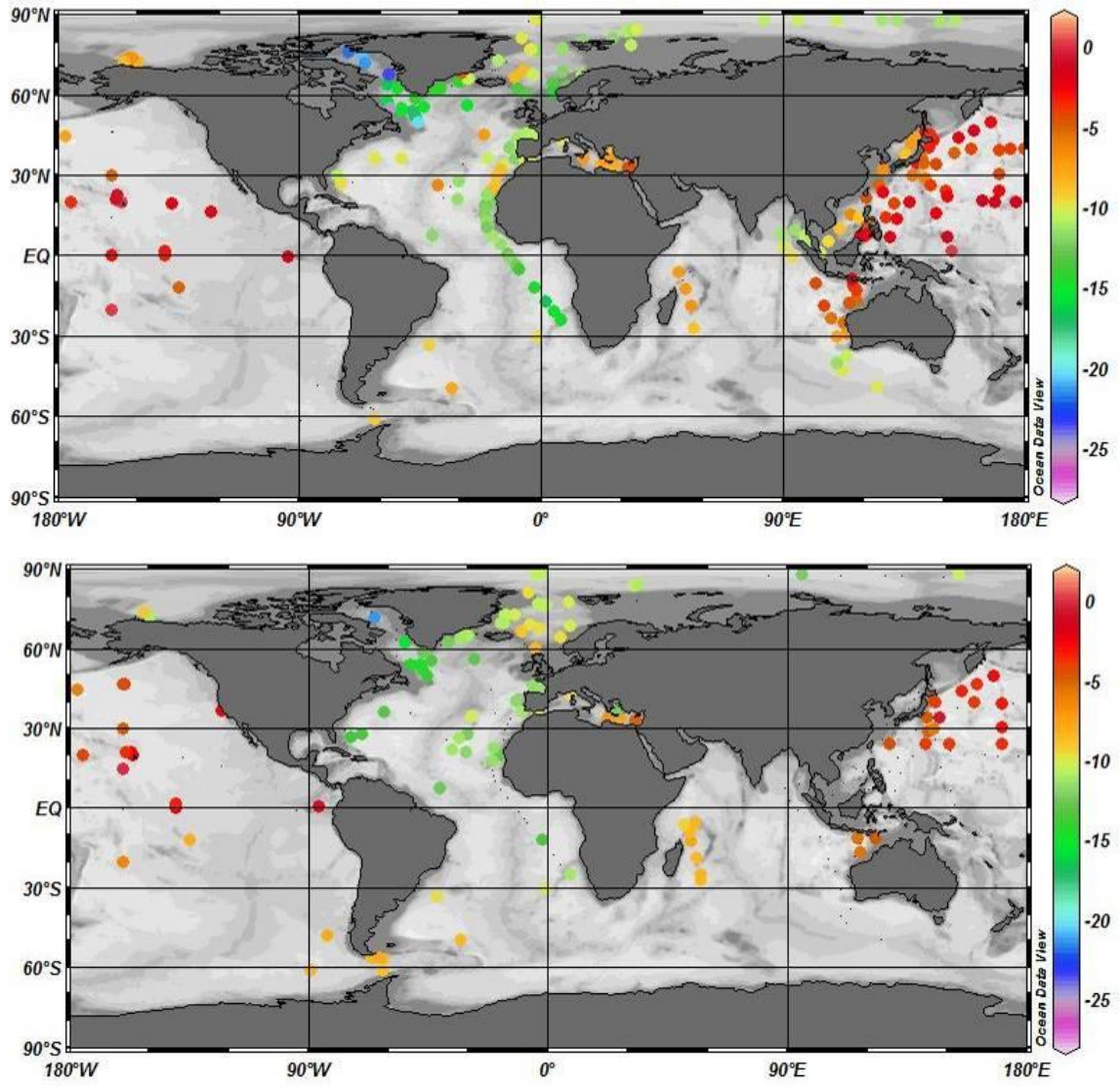


Fig. 4.
ε_{Nd} averaged between the surface and 400m depth (top) and between 800m depth and the bottom (bottom). Figure made with Ocean Data View [*Schlitzer*, 2009].

Figure 5 displays a statistical distribution of ε_{Nd} values in distinct oceanic regions. This is an updated version of a figure published in 2005 [*Lacan and Jeandel*, 2005b], with 61% more data points, and some more details. Overall, this distribution confirms the preceding one. It includes a new basin, the Arctic Ocean, which was almost not documented in 2005. The Arctic distribution is strikingly similar to that of the Nordic Seas. Again, globally, this figure illustrates the gradual ε_{Nd} increase from the northwest North Atlantic, via the Austral and Indian oceans, to the Pacific, confirming the usefulness of this tracer for studying large scale oceanic circulation.

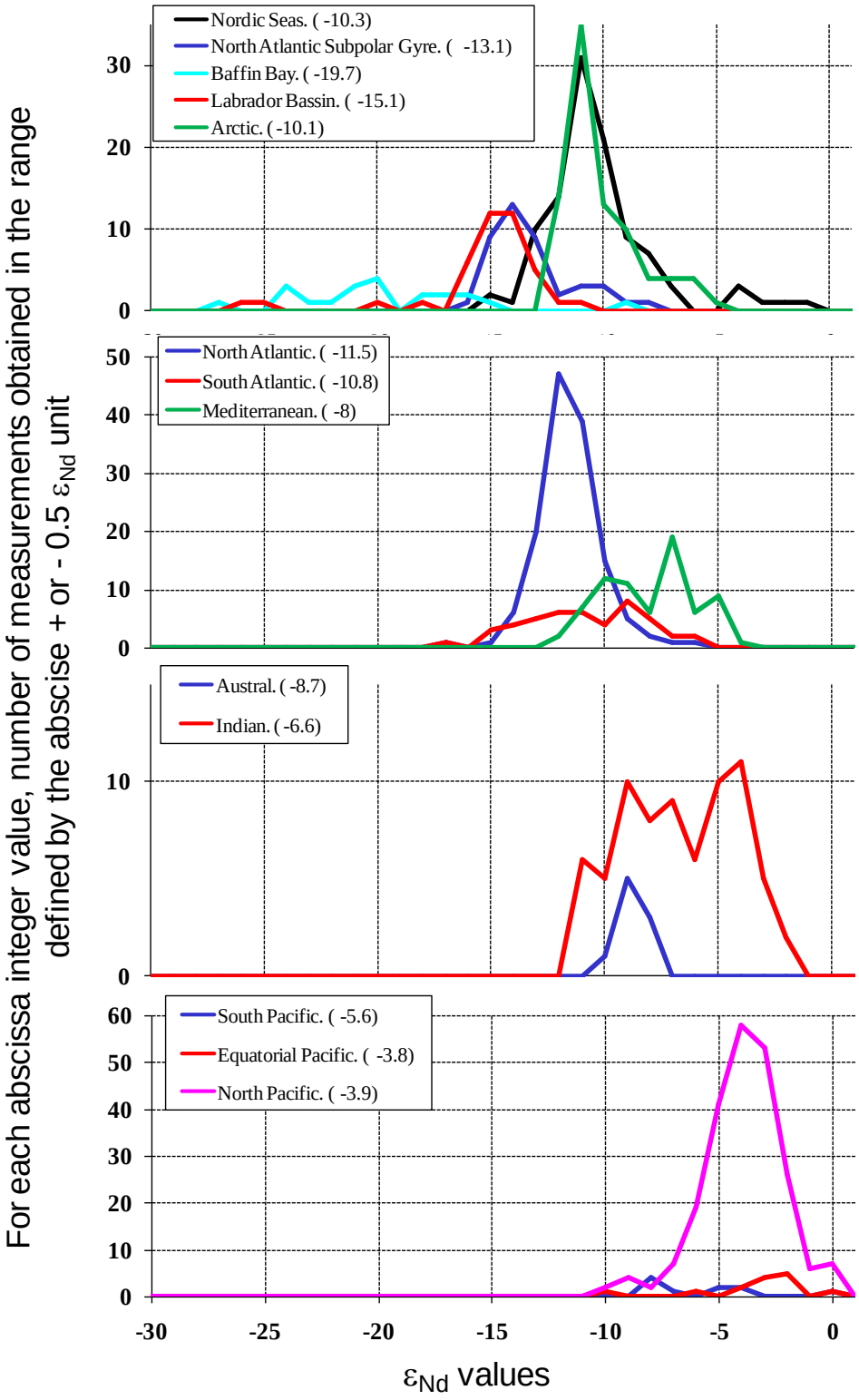


Fig. 5.
Distribution of ε_{Nd} values in distinct oceanic regions. Numbers in brackets are the average value for each region. Note that the "North Atlantic" curve excludes data from the North Atlantic Subpolar Gyre, Labrador Basin, Nordic Seas and Baffin Bay (which are plotted on the top panel).

Neodymium concentrations are displayed in Fig. 3 and Supplementary Fig. 1. Surface values are very variable. A few data points (13; less than 2% of the total number of data) display extremely high values (up to 152 pmol/kg), which likely reflect external inputs in coastal areas (e.g. Fjords). Excluding those, as a general rule, Nd concentration increases with depth. This increase contrasts with the relative homogeneity of the Nd isotopic composition (Fig. 3 and Supplementary Fig. 1). The Nd concentration vertical gradient is maximum in the Pacific, less pronounced in the Austral and Indian, much more moderated in the Atlantic, and not the dominant feature in the Mediterranean and the Arctic. The Nd concentration vertical gradient increase along the global thermohaline circulation has been attributed to the cumulative effect of particle remineralization as deep water age (as for nutrients) [*Siddall et al.*, 2008; *Arsouze et al.*, 2009]. The present compilation also suggests a possible impact of the size of each basin (i.e. ratio of margin length to basin surface area), itself being related to the intensity of external inputs to each basin. In other words, the vast Pacific Ocean is less subjected to external inputs than smaller basins (e.g. the Mediterranean and the Arctic), which likely induces the lower concentrations observed in the upper part of the water column in the Pacific than in closer basins.

Figure 6 shows the temporal distribution of the ε_{Nd} data. It shows a relatively stable and limited number of data produced in the 80s and 90s, followed by a sharp increase in the last decade. This illustrates the pioneer nature of the work conducted during these two first decades, followed a drastic increase in the interest of the oceanographic community for this tracer, which is now recognized as a core parameter of the international marine geochemical program GEOTRACES. This trend will therefore very likely go on in the framework of the GEOTRACES program. As the GEOTRACES International Data Assembly Centre will take over the management of the oceanic Nd isotope database, we will keep this pre-GEOTRACES database available on the internet. Any contribution from colleagues to correct or complement it would be appreciated.

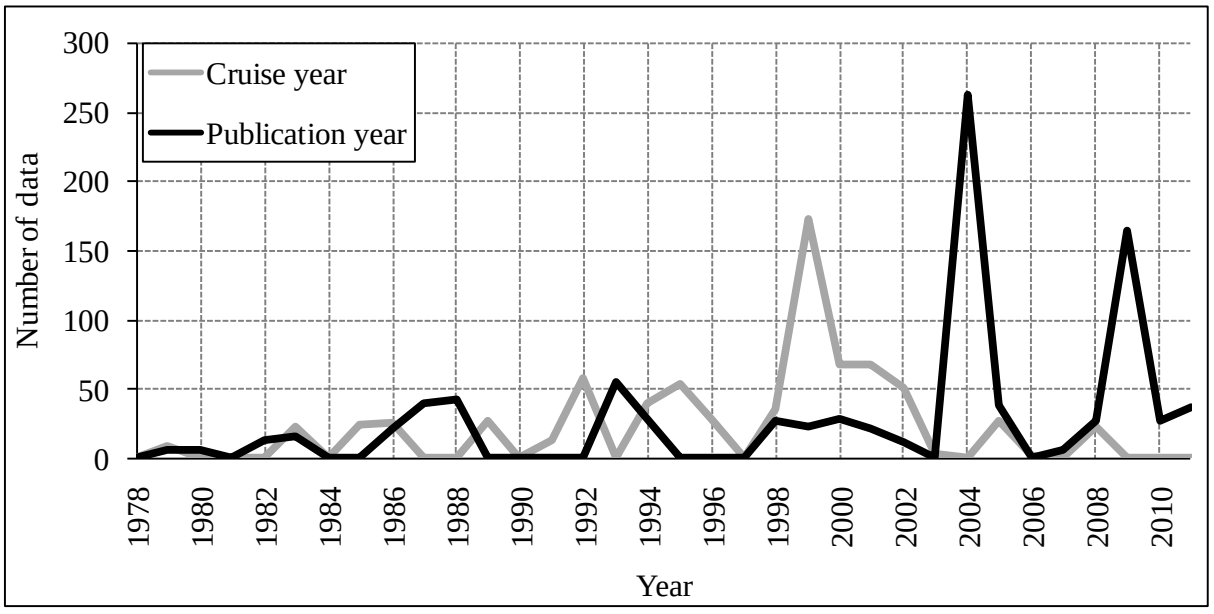


Fig. 6.
Number of ε_{Nd} data as a function of cruise and publication years. Note that cruise years are not always reported in the publications.

The geographical distribution of the data described above suggests that, in the present context, documenting the southern oceans (in particular south of 30°S) is a high priority (cf. Fig. 2b). In that

respect, major improvements are expected in the years to come, resulting from GEOTRACES and the 2007-2008 International Polar Year cruises (see the GEOTRACES International Data Assembly Centre web site for maps showing the location of these cruises).

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FIGURE CAPTIONS

Supplementary Fig. 1.

All ε_{Nd} data as a function of depth, and corresponding Nd concentration when documented, for different oceanic areas. The Nd concentration scale is restricted to values lower than 65 pmol/kg for clarity, excluding 13 shallow near shore data (< 2% of the total) from the North Atlantic. Figure made with Ocean Data View [*Schlitzer*, 2009].